Magnetic frustration effects in uranium intermetallics

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Abstract. The effect of geometrical frustration on the development of the heavy-fermion state and quantum criticality is studied in $UAuCu_4$, $UAuPt_4$, UAu_3Ni_2 samples through measurements of their magnetic susceptibility, heat capacity, and electrical resistivity. In addition, since lattice disorder can play a large role in defining magnetic properties in frustrated systems, extended X-ray absorption fine structure (EXAFS) data have also been obtained. The local structure results show a strong correlation with the magnetic properties in these samples.

Geometrical frustration often leads to a variety of interesting states of matter, such as spin-ice, spin-liquid, and spin-glass states [1, 2], which have been widely studied in pyrochlores [3] and Mott insulators [4]. In contrast, the effect of frustration on the development of the heavy-fermion (HF) state [5] or quantum criticality in intermetallic compounds [6] has received much less attention. As the class of strongly correlated intermetallics demonstrating strikingly different behavior from those of normal Fermi Liquids grows, it becomes increasingly important to account for the role of frustration in generating such non-Fermi-liquid (NFL) behavior, including the role of spin fluctuations and disorder in the quantum critical region of HF systems [7, 8, 9].

To study the effects of magnetic-order suppression and quantum criticality from frustration in HF compounds, we have studied a $UM_{5-x}X_x$ series of intermetallic compounds with the $AuBe_5$ structure: $UAuCu_4$, $UAuPt_4$ and UAu_3Ni_2. Since these *fcc* lattices have antiferromagnetic (AF) interactions, their spins are geometrically frustrated, although more weakly than that of, say, the triangular Kagomé lattice [2]. Previous measurements of the parent compounds UPt_5 and UNi_5 show standard HF behavior at low T: the linear specific heat coefficients, γ, are of order 100 mJ/mol·K^2, both the specific heats over temperature (C/T) and the magnetic susceptibilities, $\chi(T)$, go to a constant, and the dc resistivities, $\rho(T)$, follow a T^2 law [10, 11]. UCu_5 displays similar behavior, except for a magnetic transition that occurs near 1 K [12, 13]. Varying M and X atomic species should alter the electronic density and conduction-band hybridization of the f electronic states. In addition, X/M site interchange can occur, where some X and M atoms then occupy the same symmetry site, as in $UPdCu_4$ [14]. Such chemical disorder can generate random bond length disorder, leading to magnetic disorder, the effects of which would be reflected in transport and magnetic properties. In this paper, we report magnetic and transport properties for this family of intermetallics, correlated with local structural measurements using the extended X-ray absorption fine-structure (EXAFS) technique.

Samples of $UAuCu_4$, $UAuPt_4$, and UAu_3Ni_2 were arc-melted on a water-cooled copper hearth

Table 1. (Top) Summary of magnetic, specific heat data, and EXAFS fit results for three compounds. R_{nom} is the shortest U-M pair distance from the nominal crystal structure at room temperature. Note: in the fit for γ of $UAuCu_4$, we only used the data below $T = 30$ K.

	γ (J/mol·K^2)	μ_{eff} (μ_B)	T_N or T_f (K)	θ_{CW} (K)	f	f_{4c}^M
$UAuCu_4$	0.1	3.19	~ 30	-161	5	0
$UAuPt_4$	0.26	3.20	$\leq$0.15	-154	>1000	5±3%
UAu_3Ni_2	0.27	3.50	~ 3.6	-150	42	20±10%

U-M(16e)	R / R_{nom} (Å)	σ^2_{stat}($\times 10^{-4}$ Å^2)	θ_{cD}(K)
U-Cu	2.94 / 2.93	-1(1)	284(4)
U-Pt	3.09 / 3.09	6(2)	186(6)
U-Ni	2.95 / 3.02	139(2)	242(4)

with a Zr-getterered UHP Ar atmosphere, and χ, C/T and ρ were measured (figure 1). $\chi(T)$ data were fit with a Curie-Weiss law, $\chi = \frac{N_A \mu_{eff}^2}{3k_B(T-\theta_{CW})}$, above ~150 K yielding the values of μ_{eff} and θ_{CW} listed in table 1. The C/T data were fit with $C/T = \gamma + \beta T^2$ to obtain the specific heat Sommerfeld coefficient γ, with fit ranges between 2 - 30 K ($\leq T_N$) for $UAuCu_4$ and between 6 - 13 K (10 - 20 K) for $UAuPt_4$ (UAu_3Ni_2). $\rho(T)$ data at low-T were fit with a power law, $\rho = \rho_0 + AT^\alpha$, to obtain the temperature exponent, α. The frustration parameter ($f = \theta_{CW}/T_N$ or T_f, defined below) then quantifies the degree of frustration in these compounds. These properties are summarized in table 1.

X-ray absorption data were collected at U L_{III}, Au L_{III}, Pt L_{III}, Cu K and Ni K edges between 30 and 300 K on beamline 4-1 of the Stanford Synchrotron Radiation Lightsource (SSRL), using a half-tuned double crystal Si(220) monochromator with a slit height of 0.7 mm. Data were reduced using standard procedures [15, 16]. A similar set of constraints were employed to fit the data as used previously [14], which include the site-interchange model. From the fit, we get the pair distance, R, the percentage of 4c sites occupied by M, f_{4c}^M, and the Debye-Waller factor, $\sigma^2(T)$, which is then fit with the correlated-Debye model [17] using a static bond distribution width offset, σ^2_{stat}, and correlated-Debye temperature, θ_{cD} (table 1). Examples of the EXAFS data and fits are shown in figure 1d. $UAuCu_4$ data are similar to $UAuPt_4$.

$UAuCu_4$ is AF below $T_N \sim 30$ K and has a much larger $\theta_{CW} \sim$ -161 K [18]. The $\chi(T)$ and C/T data are similar to those of both its parent compound, UCu_5 ($T_N \sim 15$ K, $\theta_{CW} \sim$ -180 K) and the Ag-substituted compound, $UAgCu_4$ ($T_N \sim 18$ K, $\theta_{CW} \sim 160$ K) [18]. However, their θ_{CW} to T_N ratio are larger than for typical HF metals, such as $YbAgCu_4$ ($\theta_{CW} \sim$ -16 K and $T_K \sim 150$ K [19]), suggesting that the large θ_{CW} in the present compounds may be influenced by a large interatomic AF coupling. As $T \to 0$ K, χ and $C/T \to$ constant, consistent with Fermi-liquid behavior. The EXAFS results indicate similar local and average (nominal) structures, and the local bond length disorder, σ^2_{stat}, is consistent with zero. These data also indicate no Au/Cu site-interchange, and hence, no chemical disorder occurs, in contrast to $UPdCu_4$ [14]. All of these data are consistent with an ordered lattice and moderate AF frustration.

$UAuPt_4$, however, has quite different magnetic and transport properties. For instance, γ is more than twice that of $UAuCu_4$, $\chi(T)$ is logarithmic with T below $T \sim 20$ K, and $\rho(T) = \rho_0 + AT^{1.5}$ between $T = 0.4$ and 4 K (figure 1c inset). Even more surprisingly, C/T at $T \leq 6$ K increases linearly up to 0.65 J/mol·K^2 at 0.4 K. This behavior, given the small field dependence of C/T (not shown), cannot be described with a nuclear Schottky term or by the Hertz-Millis [20] or any other known NFL theory [21]. It is unlikely that this unusual behavior arises from a putative af transition as C/T does not show any magnetic order down to 0.15 K

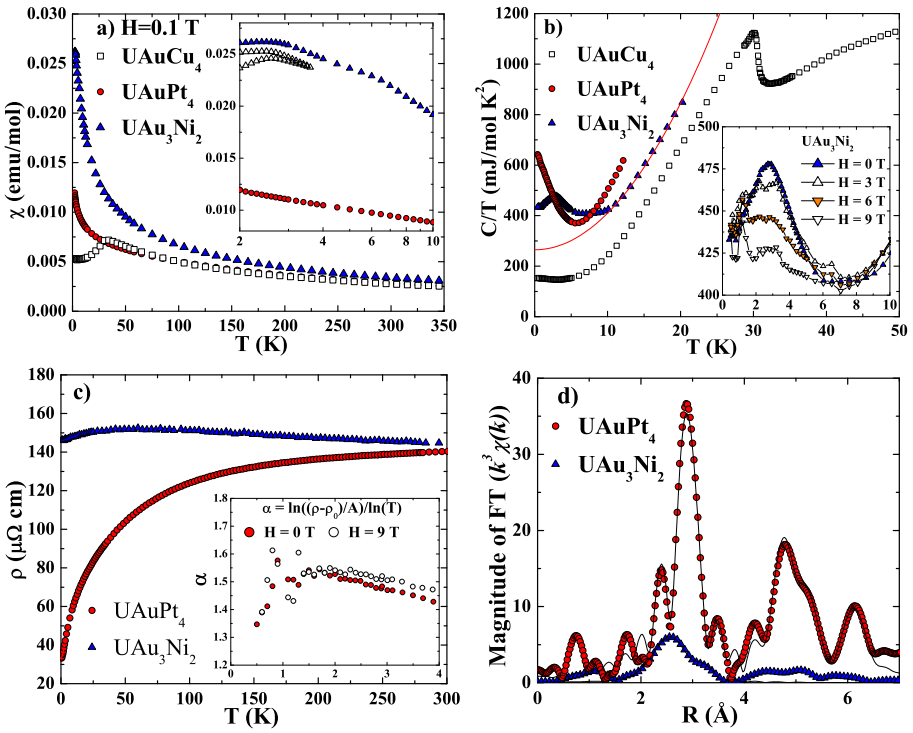

Figure 1. a) $\chi(T)$ at $H = 0.1$ T for $UAuCu_4$, $UAuPt_4$, and UAu_3Ni_2; inset shows the data below $T = 20$ K on a logarithmic temperature scale, in which the field-cooled (FC, open triangle, upper) and zero-field cooled (ZFC, open triangle, lower) data at $H = 0.01$ T for UAu_3Ni_2 are also displayed. b) C/T for three samples; inset shows the H-dependence of UAu_3Ni_2. c) ρ for $UAuPt_4$ and UAu_3Ni_2; inset shows resistivity exponent α vs. T at $H = 0$ T and $H = 9$ T for $UAuPt_4$ (open circle). d) Magnitude of Fourier transform (FT) of $k^3\chi(k)$ of U $L_{\rm III}$-edge for $UAuPt_4$ ($T = 50$ K, transformed from 3.0-15.0 Å^{-1}, and Gaussian broadened by 0.3 Å^{-1}), and UAu_3Ni_2 ($T = 30$ K, tranformed from 3.0-11.5 Å^{-1}, and Gaussian broadened by 0.3 Å^{-1}); the r-space fit (solid lines) to $UAuPt_4$ data is between 2.2-6.3 Å, and for UAu_3Ni_2 data is between 1.9-3.6 Å. Note: $\chi(k)$ represents the EXAFS oscillations, not the magnetic susceptibility χ.

[10]. All of these properties represent deviations from normal Fermi-Liquid behavior, suggesting a NFL ground state of some kind. Although $UAuPt_4$ has a large frustration parameter, the EXAFS data are consistent with a well-ordered lattice structure (table 1). Hence, $UAuPt_4$ appears to be a good candidate for a frustrated NFL metal.

UAu_3Ni_2, on the other hand, shows a logarithmic T-dependence of $\chi(T)$ below 20 K. At about $T_f = 3.6$ K, the zero-field-cooled (ZFC) and field-cooled (FC) $\chi(T)$ data start to diverge, indicative of a spin glass (SG). C/T of UAu_3Ni_2 has a broad peak at $T \sim T_f$ which decreases with external magnetic field, and is almost suppressed at $T \sim 9$ T. The $5f$ entropy at 6 K, $S_{5f} \sim 3.1$ J/mol·K, is smaller than $R\ln 2$. This behavior is consistent with the generic behavior of a classic SG, such as CuMn 0.3 at.% and $Eu_{0.4}Sr_{0.6}S$ [1]. $\rho(T) = \rho_0 + AT^{1.5}$ at low T; however, ρ_0 is much larger than that of $UAuPt_4$, possibly reflecting both the spin disorder of the SG state and a significant amount of lattice disorder/distortion due to random site occupancies. U/Au $L_{\rm III}$- and Ni K-edge EXAFS data indicate a surprisingly large fraction of the 4c site occupied by Ni, $f_{4c}^{Ni} \sim 20\% \pm 10\%$, suggesting a nearly random distribution of the Au and Ni on both 4c and 16e sites (in the nominal structure, $f_{4c}^{Ni} \sim 0$, while in a random distribution model, $f_{4c}^{Ni} \sim 40\%$).

In addition, we find a large bond static bond length disorder for the nearest-neighbor atom pair, $\sigma^2_{stat} \sim 0.0139$ Å^2. The average bond lengths also deviate from the nominal crystal structure, e.g., the distortion for the shortest U-16e pairs is R_{U-Ni} - $R_{U-Au'} \geq 0.1$ Å. The significant measured disorder is consistent with the large ρ_0 and SG-like behavior. Together with $\chi(T)$ and C/T data, we consider UAu_3Ni_2 to be a frustrated SG.

It is possible to qualitatively understand these $UM_{5-x}X_x$ intermetallics within a magnetic frustration (Q) vs. Kondo coupling (K) phase diagram, as in reference [22]. $UAuCu_4$ is well within the AF metal region with moderate frustration and weak Kondo coupling strength. Isostructural $UAuPt_4$ has very different properties without any apparent magnetic order. Since χ, C/T and ρ data all show NFL behavior above $T = 0.15$ K [10], it appears to be in the spin liquid (SL) phase. In addition, α from $\rho \propto T^\alpha$ changes very little in applied fields up to 9 T, indicative of a much larger separation from the heavy Fermi liquid (HFL) phase than $YbRh_2Si_2$ [6, 23]. However, the low-T slope in C/T decreases slightly at $H = 9$ T (not shown). The H-dependence of C/T and ρ therefore suggests that $UAuPt_4$ is near the AF and SL boundary, far away from the HFL phase, and hence close to the quantum critical point between AF and SL. UAu_3Ni_2, however, is not easily included in the two-dimensional (2D) Q vs. K phase diagram since it is also strongly disordered.

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